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Growth of ferromagnetic semiconductor: (Ga, Cr)As

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A type of GaAs-based ferromagnetic semiconductor, (Ga, Cr)As, was successfully prepared by low-temperature molecular-beam epitaxy. The (Ga, Cr)As thin film shows a flat surface up to the Cr content $x=0.18$. However, when Cr content x exceeded 0.20, the roughness of the film became larger. The ferromagnetic ordering of the semiconductor sample with the Cr content of $x=0.11$ was observed. The electronic structures of (Ga, Cr)As were characterized by x-ray absorption spectroscopy (XAS). The XAS spectra show different line shapes in different Cr content. [DOI: 10.1063/1.1455611]

I. INTRODUCTION

Because of applications to the "spintronic" devices, magnetic semiconductors have attracted much attention in recent years. Since the Mn-doped GaAs-based III–V magnetic semiconductor (Ga, Mn)As was successfully prepared,[1,2] this material has been extensively investigated.[3–6] However, the Curie temperature (T_C) of these (Ga, Mn)As has not exceeded 120 K so far, because of the solubility limit of Mn and the carrier self-compensation.[7] On the other hand, the theoretical calculation predicts that the Cr-doped GaAs shows desirable magnetic properties, because Cr $3d$-derived states in Cr-doped GaAs have a larger partial density of state near the Fermi level, which leads to a larger $p-d$ interaction than Mn $3d$-derived states in Mn-doped GaAs.[8] Up to now, although several groups have been engaged in growing Cr-doped GaAs, ferromagnetic behavior which was theoretically predicted has not been reported yet.[9] In this article, we report on a successful preparation of a GaAs-based ferromagnetic semiconductor with a high Cr content, (Ga, Cr)As, and its electronic structures and magnetic properties.

II. EXPERIMENT

The (Ga, Cr)As films were grown on an epiready GaAs(001) substrate by a conventional solid-source molecular-beam epitaxy. Reflection high-energy electron diffraction (RHEED) was used to monitor the surface structure during the growth. To fabricate the samples with different Cr content, the temperature of the Cr cell was changed from 950 to 1100 °C. After the removal of a surface oxide layer, a 150 nm GaAs buffer layer was grown at 580 °C, followed by the growth of another 150 nm low-temperature (LT)–GaAs buffer layer at 200 °C. All the growth was performed under the As stabilized condition. The first GaAs layer showed (2×4) surface reconstruction, which changed to $c(4\times4)$ when the substrate temperature (T_s) was lowered. The $c(4\times4)$ pattern was then changed to (1×1) pattern after the starting of a LT-buffer layer growth at 200 °C. The (Ga, Cr)As film was grown by opening a Cr cell during the LT–GaAs growth at 200 °C. The thickness of (Ga, Cr)As was 100 nm. After the growth of the (Ga, Cr)As film, a 20 nm GaAs cap layer was grown to prevent the oxidation of the (Ga, Cr)As film. The Cr content x in the $Ga_{1-x}Cr_xAs$ film was determined by secondary ion mass spectroscopy. The surface morphology was observed by atomic force microscopy (AFM) using the tapping mode. The magnetic properties of the samples were measured by a superconducting quantum interference device magnetometer (SQUID). To investigate the electronic structure of the (Ga, Cr)As films, x-ray absorption spectroscopy (XAS) was performed at the BL-11A of the Synchrotron Radiation Research Center (SRRC) in Taiwan. To obtain an information from the whole (Ga, Cr)As layer, XAS was measured using partial fluorescence yield using Ge detector where the fluorescence from Cr L edge was detected.

III. RESULTS AND DISCUSSION

The RHEED patterns for the (Ga, Cr)As films with different Cr content are shown in Fig. 1. The (1×1) patterns

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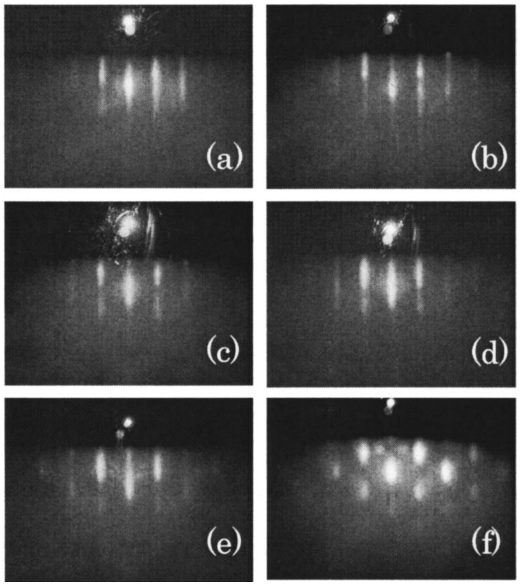

FIG. 1. Sequence of the RHEED patterns taken along the [1-10] azimuth of the (Ga, Cr)As films. Figure 1(a) corresponds the RHEED pattern before the growth of the (Ga, Cr)As films, and Figs. 1(b)–1(f) corresponds the RHEED patterns of (Ga, Cr)As films with the Cr content of $x=$ (b) 3.0%, (c) 6.6%, (d) 14%, (e) 19%, and (f) 26%, respectively.

were very streaky despite the low-temperature growth up to the Cr content $x=0.18$ [Figs. 1(b)–1(e)]. However, the pattern became spotty for the sample with the Cr content $x=0.26$ [Fig. 1(f)]. From the surface morphology observation with AFM, the surface of the sample with the Cr content $x=0.066$, which corresponds to the sample of the Fig. 1(c), was relatively flat and the rms roughness was estimated to be 1.72 nm. On the other hand, the surface of the sample with Cr content $x=0.26$, which corresponds to Fig. 1(f), became relatively rough and the rms roughness was estimated to be 2.82 nm. It is suggested that with increasing the content of Cr, which is incorporated into the films, the lattice mismatch becomes larger and the three-dimensional (3D) islands are likely to form to relax the lattice mismatch. The threshold of the formation of the 3D island seems to lie near the $x=0.20$.

The magnetic properties of semiconductor samples of (Ga, Cr)As were measured by SQUID. Figure 2(a) shows the magnetic field dependence of the magnetization of a sample with $x=0.11$ at 5 K. The magnetic field was applied parallel to the (Ga, Cr)As epitaxial film. The diamagnetic contribution of the GaAs substrate, which was measured separately, was subtracted from the total magnetization. The inset shows the magnified figure around the zero fields and a clear hysteresis loop was observed. From these results, the presence of a ferromagnetic ordering with $x=0.11$ was observed. The T_C of our sample is still much lower than room temperature. However, it is well known that magnetic properties of GaAs-based magnetic semiconductors grown at low temperature strongly depend on the growth condition.[5,6] Therefore, there is a possibility that the magnetic property of the (Ga, Cr)As will be much improved with the optimization of the growth condition and the Cr concentration. The magnetic properties of the (Ga, Cr)As will be described in detail elsewhere.[10]

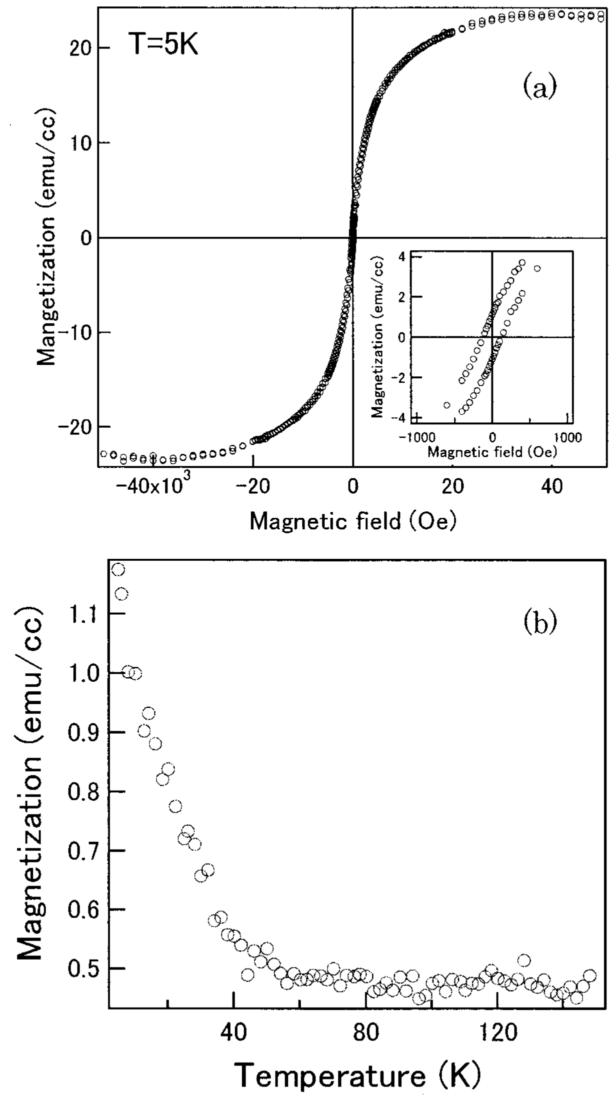

FIG. 2. (a) Magnetization of (Ga, Cr)As at 5 K with Cr=11%. Inset shows the magnified figure around the zero magnetic field. The ferromagnetic ordering was clearly observed. (b) Temperature dependence of the remanent magnetization of the same sample.

To investigate the electronic structure of (Ga, Cr)As, an XAS was performed at the Cr L edge. Figure 3 shows the XAS spectra of (Ga, Cr)As films with Cr content from $x=0.01$ to $x=0.5$. The peak width becomes wider with increasing the Cr contents. It is suggested that with increasing the Cr contents, the electronic structures of the Cr $3d$ changes and Cr $3d$ has the different electron configurations. This result corresponds to the semiconductor-to-metal transition in this system which occurs around the Cr content of $x=0.2$.[10]

IV. CONCLUSION

In summary, we have successfully grown a GaAs-based magnetic semiconductor film with high Cr doping; (Ga, Cr)As. The ferromagnetic ordering was observed with a

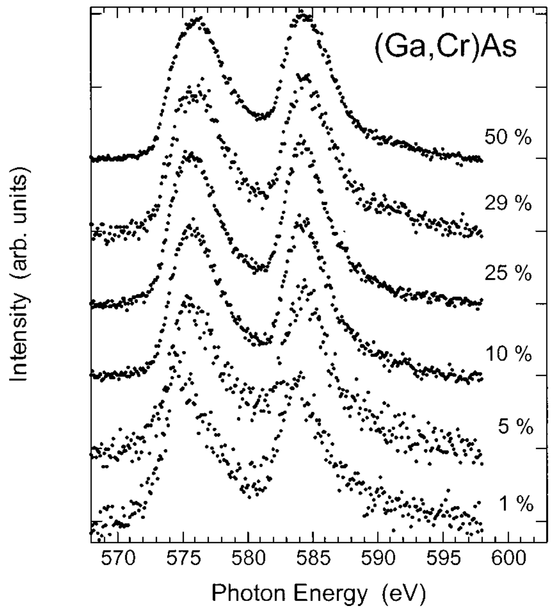

FIG. 3. XAS spectrum of (Ga, Cr)As with the Cr content from $x=0.01$ to $x=0.5$ at the Cr L edge measured by partial fluorescence yield.

clear hysteresis loop from the semiconductor sample with Cr content of $x=0.11$ using a SQUID. We believe that this material has a potential to have higher T_C. The electronic structures of the (Ga, Cr)As were studied by XAS. The change in the peak width with increasing the Cr content was observed.

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